Ultrafine Particle (UFP) Exposures in an Aluminium Smelter: Soderberg vs. Prebake Potrooms

Maximilien Debia (Corresponding author), Scott Weichenthal, Robert Tardif & André Dufresne
Department of Occupational and Environmental Health, Faculty of Medicine, Université de Montréal
P.O. Box 6128 Main Station, Montréal, Québec H3C 3J7, Canada
Tel: 1-514-343-6111 (3782) E-mail: maximilien.debia@umontreal.ca

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Abstract

The objectives of this work were to evaluate ultrafine particle (UFP) exposures during aluminium smelting in Soderberg and prebake potrooms. Particle concentrations were monitored using the P-Trak 8525 and aerosol particle size distributions were monitored with the electrical low pressure impactor (ELPI). UFP samples were analyzed for chemical composition by transmission electron microscopy (TEM). Workers who accomplished tasks in the Soderberg cell environment were more exposed to UFP than those who worked in the prebake; however, the specific task of anode shift in the prebake process was an important source of UFPs. More than 92 % and 98 % of particles had aerodynamic diameters less than 98 nm in the Soderberg and prebake processes, respectively. TEM/EDS analysis suggests that workers are exposed to short fibrous aluminium particles with a nanometric aerodynamic diameter. Overall, this study suggests that occupational hygiene practices aimed at evaluating UFP exposures should include monitoring of the particle number concentration, monitoring of the particle size distribution, and characterization of the nanoscale fraction of the aerosol.

Keywords: Ultrafine particles, Exposure assessment, Potroom, Aluminium smelter, TEM/EDS

1. Introduction

Ultrafine particles (UFPs) are generally defined as those particles with a diameter less than 100 nm which are not intentionally produced unlike the nanomaterials or nanoparticles (Preining, 1998; Brouwer, Gijsbers, & Lurvink, 2004; Pfefferkorn et al., 2010). Sources of UFPs exist in a number of industrial settings including smelters (Cunningham, Jablonski, & Todd, 1996; Thomassen et al., 2006), arc spraying (Rendall, Phillips, & Renton, 1994), gas metal arc welding (Brouwer, Gijsbers, & Lurvink, 2004; Hewett, 1995; Hovde & Raynor, 2007; Zimmer, Baron, & Biswas, 2002), high speed grinding (Zimmer & Maynard, 2002), foundries (Evans, Heitbrink, Slavin, & Peters, 2008) micro-machining processes (Handy, Jackson, Robinson, & Lafreniere, 2006), and various other job activities (Elihn & Berg, 2009). In particular, primary aluminium smelters are known to generate high concentrations of UFPs (Elihn & Berg, 2009; Thomassen et al., 2006). However, UFP concentrations (particles/cm^3) are rarely examined when assessing worker exposures in smelters and generally only exposures such as inhaled mass dusts, fluorides, coal tar pitch volatiles, Polycyclic Aromatic Hydrocarbons (PAHs) and sulfur dioxide (SO_2) are monitored (Benke, Abramson, & Sim, 1998). Abramson et al. (2010) recently reported a clear dose-response relationship between cumulative exposures to SO_2, fluoride, inhalable dust, and the benzene soluble fraction at levels below the exposure standards and pulmonary effects such as asthma symptoms, airflow limitation, longitudinal decline in lung function, and non-specific bronchial hyper-responsiveness among aluminium smelter workers. However, UFPs were not examined in this study and Abramson et al. (2010) indicated that other constituents of inhalable dust could be responsible for at least a portion of potroom asthma. Since the prevalence of occupational UFP exposures may be high in potrooms, it is important to identify the determinants of this exposure. Indeed, this issue is of increasing importance as evidence suggests that UFPs are potent triggers of oxidative stress and may contribute to adverse respiratory and cardiovascular outcomes (Donaldson, Stone, Gilmour, Brown, & MacNee, 2000; Oberdorster, Oberdorster, & Oberdorster, 2005; Oesterling et al., 2008; Sioutas, Delfino, & Singh, 2005; Warheit, Sayes, Reed, & Swain, 2008; Wittmaack, 2007).

There are two major types of Hall-Heroult electrolytic cells in use in aluminium smelters: prebake and Soderberg processes. The prebake process utilizes manufactured electrodes in an “enclosed” process whereas Soderberg-type cells use a mixture of coke and pitch binder that is put into pots and immersed in an “opened” molten bath mixture. In most industrial countries, the Soderberg process has been replaced by the prebake process as it generates fewer emissions. For UFPs specifically, the task of anode shift in prebake type cells has been identified as an important source of exposure (Thomassen et al., 2006). However, further investigation is required to characterize UFP levels in specific work locations in order to better characterize potential occupational health risks. To address this need, the current study was conducted with the following objectives: 1) measurement of UFP concentrations produced during aluminium smelting in Soderberg and prebake potrooms; 2) estimation of workers’ exposures in specific job tasks; 3) evaluation of the size distribution of UFPs produced during aluminium smelting and; 4) characterization of the chemical composition of the smallest aerosol size fractions.

2. Methods

2.1 Aluminium Smelting Reduction Cells

Two aluminium smelting reduction cells were studied in a large aluminium production facility of the Province of Québec. The first process was the Soderberg electrolytic reduction cell which was composed of two potrooms of approximately 180 smelting pots. The second process was the prebake reduction cell composed of two potrooms of approximately 240 pots. The two processes were located in the same building but were separated by physical barriers and separate exhaust ventilation systems, and as such were considered isolated.

2.2 Instrumentation

UFP number concentrations were monitored using four TSI P-Trak 8525 ultrafine particle counters (TSI Inc.) at 10 second sampling intervals. These instruments use a laser light source and optical sensor to count particles after air has passed through a chamber saturated with isopropyl alcohol. Aerosol particle size distributions were monitored in real-time with an electrical low pressure impactor (ELPI) with 12 impactor stages (ELPI, Dekati Ltd., Tampere, Finland). The ELPI measures airborne particle size distributions in the range of nanometers. The electrical detection of aerosol particles is made in a low-pressure cascade impactor where the pressure under the first stage is adjusted to 100 mbar. Two ELPI configurations were used. First, aerosol particle size distributions were monitored using sintered collection plates greased with 1-2 oil droplets. According to this setting, the cut-points (in µm) of the 12 impactor stages were: 0.024, 0.030, 0.050, 0.098, 0.214, 0.321, 0.583, 0.902, 1.524, 2.272, 3.796 and 6.351 (Dekati, 2006). Second, aerosol samples were collected on polycarbonate filters and the five lower stages of the low-pressure impactor were subsequently analyzed for chemical composition by transmission electron microscopy (TEM) coupled with an energy dispersive X-ray spectroscopy (EDS). Substrates were not greased in this procedure. According to this setting, the cut-points (in µm) of the 12 impactor stages were: 0.028, 0.055, 0.093, 0.156, 0.263, 0.384, 0.616, 0.953, 1.61, 2.40, 4.01 and 6.71 (Dekati, 2006). Copper-grids (200 mesh) coated with carbon film (which contains fraction of Si) were stuck directly on the substrates using glue. Only a fine part of the grid was stuck.

2.3 Monitoring Strategies

A systematic mapping of UFPs was performed in the general work environment of each potroom and “quasi-personal” exposure measurements were also monitored for different job activities using P-Trak instruments. Mapping was carried out in two Soderberg electrolytic reduction cell potrooms and in two prebake reduction cell potrooms. Direct measurements were made with a P-Trak in front of each pot under real-time working conditions. In the Soderberg potrooms, measurements were performed at a distance of 2 meters from the pots with each separated by a distance of approximately 15 meters. In the prebake potrooms, measurements were performed 2 meters from the pots with each separated by a distance of approximately 5 meters.

“Quasi-personal” exposure measurements were collected as close as possible to workers’ breathing zones while they were performing their tasks. Sampled work-tasks included maintenance of the pots and breaking the crust in the Soderberg reduction cell potrooms. The maintenance labour is assigned to the anodes sealing which consists of pushing the alumina near the electrolytic bath whereas the job of the labour assigned to break the crust consists of putting a wood stick in the electrolytic bath in order to drill a gas pocket. Sampling time for these tasks ranged from one to two hours. For truck drivers and overhead bridge crane operators, P-Trak instruments were placed in the vehicle cab and measurements were performed over 2-4 hour periods. Tasks monitored for truck drivers included replenishing cells with briquettes and adjusting steel studs immersed in the anode, only in Soderberg potrooms. Tasks monitored for overhead crane operators included anode shift and distribution of alumina in prebake type cells. Sampling times represent the specific work between breaks which is representative of a whole working day. At each break, P-trak instruments were collected and used to estimate "quasi-personal" exposures of another worker. Measurements were repeated during four consecutive working days. The monitoring strategy was developed as close to random as possible.

Aerosol particle size distributions were monitored in real-time with the ELPI at a distance of 1-2 meters from the pots over a period of 10 to 20 minutes. The collection of samples for TEM analyses was performed over a 7 minute period for the Soderberg process and a 10 minute period for the prebake process as not to overload TEM grids.

2.4 TEM Analyses

TEM grids were analyzed using a JEOL microscope (model JEM-2100F) equipped with a Field Emission Gun (FEG) running at 200 kV. Particle images were taken directly on the grids without any further treatment. Chemical composition was determined for 93 and 77 particles in the Soderberg and prebake potrooms, respectively and was carried out using EDS with a detection limit of 0.3 % weight (3000 ppm). Only particles deposited on the five smallest stages of the impactor were analysed by TEM/EDS.

3. Results

3.1 Particle Number Concentrations (mapping)

Outdoor background concentrations at the site were lower than 7000 particles/cm³. Average UFP concentrations were 144 000 particles/cm³ in the Soderberg potroom and 70 000 particles/cm³ in the prebake potroom (Table 1). Figure 1 presents the relationship between the mapping UFP concentrations of the Soderberg and Prebake potrooms. The geometric mean (full line) of Soderberg concentrations was approximately three-fold greater than for the prebake. The 5th-95th percentiles boxes of the log-transformed distributions indicate large variation in particle number for the prebake process. The geometric standard deviations (GSD) calculated for the two processes were 1.7 for the Soderberg process and 2.6 for prebake, respectively.

3.2 Quasi-personal Sampling

UFP exposure data for various job activities are summarized in Table 1. In general, exposures for overhead crane operators were significantly lower than for other job activities for both processes. However, anode truck drivers had the lowest monitoring concentrations (GM = 14 000 particles/cm³) of the tasks monitored in the prebake process. Personal exposures tended to be greater for workers in Soderberg potrooms with the highest exposures observed for the breaking crust labour (GM = 178 000 particles/cm³) followed by the maintenance labour (GM = 113 000 particles/cm³) and the truck driver adjusting the steel studs (97 000 particles/cm³).

3.3 Aerosol Particle Size Distributions

The percentage of particles on each of the 12-stages of the ELPI impactor is presented in Table 2. Average aerodynamic diameters for particles collected in Soderberg and prebake potrooms were below 0.030 µm whereas average aerodynamic diameters of particles in prebake potrooms tended to be smaller (< 0.024 µm). Moreover, 97 % and 98 % of particles had aerodynamic diameters smaller than 0.098 µm in the Soderberg and prebake processes, respectively. Similar findings were observed when the ELPI monitoring was conducted using collection plates with polycarbonate substrate (Table 3). Specifically, 91 % and 90 % of particles had aerodynamic diameters below 0.093 µm in the Soderberg and prebake processes, respectively, with average aerodynamic diameters below 0.028 µm for both processes.

3.4 Chemical Characterization by TEM/EDS

The results of TEM/EDS analysis are shown in Table 4. The elements detected are presented in decreasing order for the fifth lowest stage of the ELPI. In general, Al, Na and F were the most common elements detected. Moreover, particles containing Al, Na and F were dominant in both the prebake and Soderberg processes with relative abundances of approximately 69 % and 54 %, respectively. Particles containing Ti were also prevalent in the workroom air with relative abundances of approximately 9 % (prebake) and 33 % (Soderberg), respectively. Moreover, 25 % (prebake) and 31 % (Soderberg) of particles examined were fibres and were found on the fifth lowest stage of the ELPI for the two processes. A representative aluminum fibre from the prebake process is shown in Figure 2 and contained primarily Al and Na (C, Si, O and Cu peaks are specific to the substrate and the grid).

4. Discussion

4.1 Particle Number Concentrations (mapping)

An evaluation of exposures to UFPs in the potlines of an aluminum reduction plant was performed in both the Soderberg and prebake processes. UFP concentrations in the Soderberg process exceeded those in prebake potrooms; however, peaks were observed in the prebake process when the doors of the pot were opened by maintenance labour and when the anode was removed from a pot by the crane operator and was left in the aisle to cool. In general, UFP emissions in the Soderberg process were relatively stable whereas emissions from the prepake process tended to be more sporadic. Similar observations were reported by Thomassen et al. (2006) who suggested that the specific task of anode shift in the prepake process is an important determinant of UFP concentrations in potrooms.

4.2 Quasi-personal Sampling

The highest particle number concentrations were observed among workers who had to break the crust of the electrolyte bath of the Soderberg cell. This task, performed manually, generates large quantities of fumes and workers were close to emission sources most of the time. Gylseth, Bjørseth, Dugstad, & Gjønnes (1984) indicated also that fibrous particles were found in high concentrations during this specific operation. The second highest exposures were for maintenance workers. These workers continuously follow the crane operator and perform all of their tasks in the potlines. Although general background UFP levels were notably lower in the prebake process, maintenance workers had to open the doors of the enclosed pots to permit access to the crane operator who had to move or remove the anodes. However, workers wear full face powered air purifying respirators at all times during this task.

4.3 Aerosol Particle Size Distributions

In general, particles generated in the Soderberg process were slightly larger than in the prebake but more than 90 % of particles in both processes were in the ultrafine range (< 100 nm). These results are in concordance with other studies which have monitored aerosol size distribution using scanning mobility particle sizers (SMPS) (Thomassen et al., 2006; Elihn & Berg, 2009). Specifically, Thomassen et al. (2006) reported size distribution function peaks around 40 nm in the Soderberg process and below 20 nm in the prebake. However, in their study, Thomassen et al. (2006) collected samples close to open cells during anode change whereas our samples were collected when no anode shift was carried out in the potroom. Our results suggest that the majority of particles generated in prebake potrooms are in the ultrafine range with or without anode work but more attention should be given to the anode change processes as high UFP concentrations were monitored during this task. Moreover, high UFP concentrations were also observed in the room where old anodes were cooling down with concentrations reaching 500 000 particles/cm³ (results not shown). Therefore, UFP exposures are also a concern in this location and worker protection should be equivalent in the two rooms.

4.4 Chemical Characterization by TEM/EDS

Particles identified by the TEM/EDS analysis were not representative of the all particles because only the fifth smallest fractions of the ELPI were studied and because C, Si, O, Cu-based particles were not detected in the EDS. Nevertheless, the morphology and chemical composition of particles were quite similar to the types of particles reported by Gylseth, Bjørseth, Dugstad, & Gjønnes (1984), Höflich et al. (2005) and Thomassen et al. (2006). Chemical characterization by EDS showed that particles containing Al, Na and F were dominant with relative abundances of approximately 69 % (prebake) and 54 % (Soderberg). These results are in concordance with the study of Höflich et al. (2005) who indicated that sodium β-alumina ($NaAl_{11}O_{17}$) and cryolite (Na_3AlF_6) were dominant oxide and fluoride present in workroom air of aluminum smelter potrooms.

The specific fibre shown in figure 3 was collected in the prebake potroom while no activity was carried out, suggesting that workers were exposed to these fibres in all the prebake potrooms, with or without anode work. This finding is also a concern since worker protection is not mandatory in the location without anode work. Gylseth, Bjørseth, Dugstad, & Gjønnes (1984) reported high concentrations of fibrous sodium aluminumtetrafluoride rangeing from 9 to 720 fibers/cc in potrooms of the prebake and Soderberg processes. The fibres were described as thinner than 0.1 µm in diameter and shorter than 5 µm. Voisin et al. (1996) confirmed the presence of short fibrous aluminium particles (mean length of 1 to 2 µm), in the bronchoalveolar lavage fluid from four primary aluminium workers and considered them as various forms of aluminium oxides. Biopersistence of these fibres in the respiratory tract were also suggested by Voisin et al. (1996) since fibres were identified in biological samples collected more than four years after the cessation of exposure. The present study suggests that these fibres can have an aerodynamic diameter in the ultrafine range since several fibres were identified in the lowest stage of the impactor (figure 2). The fractions of fibres in the prebake and Soderberg potrooms of 25 % and 31 %, respectively, are consistent with previous studies where the fraction of fibres was approximately 30 % of total particle number (Thomassen et al., 2006).

Another element that has attracted particular interest in this study is Ti which was found in the fifth smallest stages of the ELPI for both processes. Höflich et al. (2005) also identified Ti-oxide particles in their characterization of the workroom air of aluminum smelter potrooms. However, the relative abundances of these particles were significantly lower (0.6 % and 0.8 % for Soderberg and prebake processes, respectively) than the abundances observed in this study. This difference may be explained by the fact that only the fifth smallest fractions of the ELPI were examined by TEM/EDS. In fact, to our knowledge this is the first time that chemical characterization by TEM/EDS has been conducted for the ultrafine fraction using an ELPI impactor. Alternatively, this difference may be explained by the relatively small number of particles examined (approximately 170 particles) in both processes and new investigations are required to confirm these results.

4.5 Evaluation of UFP Exposures

There is currently no consensus on how to measure worker exposures to UFPs. In a field comparison study, Zhu, Yu, Kuhn, & Hinds (2006) concluded that the P-Trak worked reasonably well compared with other "non portable" condensation particle counters (CPC) but that caution must be given in interpreting data collected by P-Trak monitors near combustion sources. Park, Ramachandran, Raynor, Eberly, & Olson Jr (2010) indicated also that at high concentrations significant underestimation (by as much as a factor of 3) can occur when using the P-trak. Two instruments, the ELPI and P-Track, were used in the present study to determine particle number concentrations and aerosol particle size distribution. The P-trak is based on the optical properties of particles and it measures particle number from 20 nm to about 1 µm. The ELPI measures the size distribution of particles as a function of aerodynamic particle size in real-time. Using sintered collection plates configuration, the cut-point of the lowest stage is 0.024 µm. In the present study, the filter stage which permits measuring particle cut size of 7 nm was not used and the lowest cut size used was 24 nm which was near the P-trak cut size of 20 nm. Since it has been demonstrated that the majority of particles in potrooms are on the nanometer size range, data obtained with the ELPI or with the P-trak are probably underestimated. However, because the majority of particles are in the ultrafine range in potrooms, the particle number concentration obtained with a P-trak can be used as a surrogate of total UFP count number.

Several instruments can determine the particle size distribution of UFPs. ELPI and SMPS are the most popular tools and both have advantages and disadvantages. The ELPI has good time resolution but the size resolution of the SMPS is better. EEPS (Engine Exhaust particles Sizer) is a most recent tool which has, like the SMPS, a good size resolution and, like the ELPI, a good time resolution. Since a good agreement was reported between the ELPI and SMPS (Marjamaki, Keskinen, Chen, & Pui, 2000) and between the ELPI and EEPS for real-time particle size measurements (Zervas & Dorlhène, 2006), the three devices can be used by hygienists to estimate the fraction of particles smaller than 100 nm. Nevertheless, the ELPI is the only instrument which permits to collect samples of aerosol by selecting them using aerodynamic diameters. Overall, as mentioned by others authors, these instruments are still expensive and the field evaluation is also a complex process (Tsuji et al., 2006).

Sioutas, Delfino, & Singh (2005) indicated that it is essential to assess the nature and levels of UFPs to which people are exposed before undertaking epidemiologic investigations on their health effect. Thus, systematic evaluations of UFP exposures similar to the one presented or others (Elihn & Berg, 2009; Thomassen et al., 2006) are important to perform and should include monitoring of the particle number concentration, evaluation of the particle size distribution in order to calculate right UFP concentrations and characterization of UFPs by TEM/EDS.

5. Conclusion

Both Soderberg and prebake processes generate high UFPs level. However, in general, higher levels were observed during the Soderberg process. Workers who conducted tasks in the Soderberg cell environment were more exposed to UFP than those who worked in the prebake but the specific task of anode shift in the prebake process was an important source of UFPs. Since stratification of level of exposure among workers is easily possible with P-trak, it represents a useful tool for performing occupational evaluation and control of worker exposures to UFPs. TEM/EDS analysis of particles did not show differences in particle composition between the two processes but indicated that workers are exposed to nanoscale particles that contain Al, Na, F, and Ti and to nanometric aluminium fibers.

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Table 1. Descriptive statistics of particle concentrations (number/cm^3) monitoring using P-trak in the Soderberg and prebake potrooms

Process	Soderberg				
Mapping mean concentration (GSD)	144 000 (1.7) (n=137)				
“Quasi-personal” sampling - Job activity	Operator pressurized overhead bridge crane	Maintenance labour	Labour-breaking crust	Adjusting steel studs truck driver	Briquettes truck driver
N: number of samples	4	11	3	5	4
Mean	16 000	123 000	186 000	100 000	67 000
Maximum	22 000	209 000	251 000	131 000	159 000
Minimum	14 000	53 000	125 000	70 000	14 000
Geometric mean	16 000	113 000	178 000	97 000	47 000
Geometric standard deviation	1.2	2.0	1.4	1.3	2.7

Process	Prebake			
Mapping mean concentration (GSD)	70 000 (2.6) (n=205)			
"Quasi-personal" sampling - Job activity	Operator pressurized overhead bridge crane	Maintenance labour	General maintenance truck driver	Anode truck driver
N: number of samples	3	12	6	5
Mean	29 000	99 000	74 000	17 000
Maximum	46 000	217 000	113 000	31 000
Minimum	10 000	16 000	28 000	6000
Geometric mean	24 000	83 000	66 000	14 000
Geometric standard deviation	2.2	2.0	1.7	1.9

* Outdoor background concentrations at the site were lower than 7000 particles/cm^3.

Table 2. Fractions (number percents) of particles on the 12 stages of the ELPI impactor for the Soderberg and prebake processes when using sintered plates

Stages cut-points in μm	Percentage of particles in each stages of the ELPI	
	Soderberg	Prebake
0.024	42	77
0.03	32	18
0.05	19	3
0.098	4	<1
0.214	1	<1
0.321	<1	<1
0.583	<1	<1
0.902	<1	<1
1.524	<1	<1
2.272	<1	<1
3.796	<1	<1
6.351	<1	<1

Table 3. Fractions (number percents) of particles on the 12 stages of the ELPI impactor for the Soderberg and prebake processes using collection plates (polycarbonate filters)

Stages cut-points in µm	Percentage of particles on each stage	
	Soderberg	Prebake
0.028	79	69
0.055	12	21
0.093	6	7
0.156	2	2
0.263	<1	<1
0.384	<1	<1
0.616	<1	<1
0.953	<1	<1
1.61	<1	<1
2.40	<1	<1
4.01	<1	<1
6.71	<1	<1

Table 4. Major elements detected in the TEM/EDS ranking in decreasing order

	ELPI stages, cut-points in µm				
	0.028	0.055	0.093	0.156	0.263
Soderberg	Ti, Fe, Al, Na, S, K, F, Ca	Al, Na, K, Ti, S, Fe, Ca, F	Ti, S, Fe, Na, Al, K, Na, Ca	Al, Na, F, K, S, Cl, Ti, As	Al, Na, F, S, K, Fe, Ti, As, Cl, V, Au, Pb, Zn
Prebake	Al, Na, F, K, S, Fe, Mn, Ti	Al, Na, F, K, S, Ti, As	Al, Na, F, K, S, Fe, Ti	Al, Na, F, K, Ti, S, Fe, Mn	Na, Al, F, K, S, Ti, As, Cl

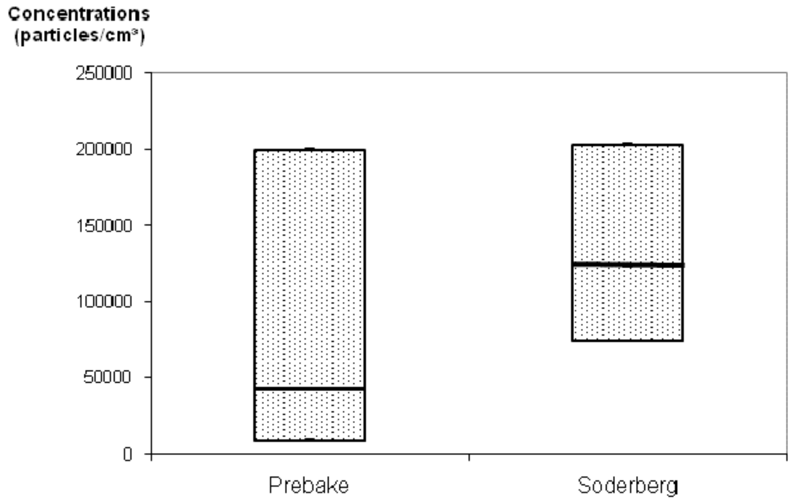

Figure 1. Relation between the mapping UFP concentrations of the Soderberg and Prebake potrooms monitoring using P-trak and shown as geometric mean (full line) and 5th-95th percentiles (box) of the log-transformed distribution

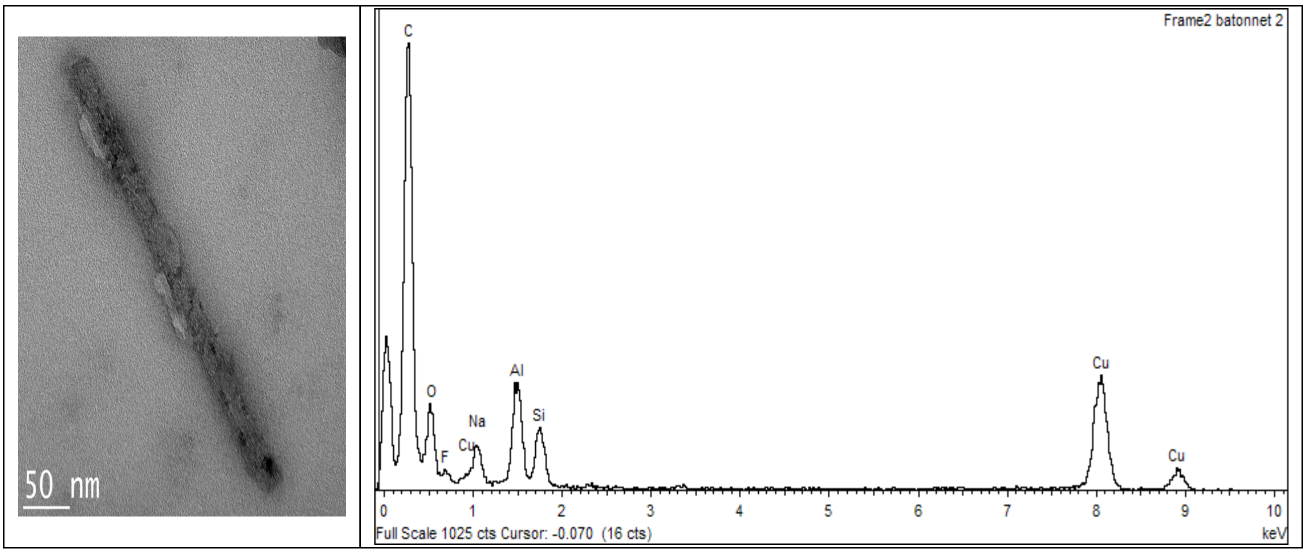

Figure 2. TEM/EDS analysis of a typical fiber particle of the prebake process on the stage 0.028 µm of the ELPI